Application of combined load for obtaining ultra-fine grained structure in magnetic alloys of the Fe-Cr-Co system

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ABSTRACT

Fe-Cr-Co system magnets are known for their high remanence and maximum energy product along with high mechanical properties. However, since the thermomagnetic treatment of the alloy implies the spinodal decomposition, which in turn drops the ductility of the material, finding a balance of magnetic and mechanical properties is in focus of many scientist due to its relevance. One of possible paths for finding this balance is application of hot deformation approach. The processes of dynamic recrystallization during hot deformation by means of compression accompanied torsion of magnetic alloys Fe-25%Cr-15%Co and Fe-30%Cr-8%Co of Fe-Cr-Co ternary system were studied. It is shown that the chosen method of deformation can be effectively applied to receive ultrafine-grained structure in the vicinity of the deformation zones and obtaining the gradient type structure in considered alloys. Founding on the analysis of results obtained, basic principles for enhancement of the alloy properties during thermomechanical treatment were figured out. Specific values of strain temperature and velocity for both considered alloys were proposed.

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1.Introduction

Generally magnetic materials play a great role in the overall production and creating of new types of magnets with elevated properties always been a challenging task of the mateiral science and related fields. However, within this large group of materials there exist several directions where the requirements to not only magetic but also mechanical properties of the magnetic materials are very high and difficult to achieve (Delette, 2015; Y. Zhang et al., 2014). Among several large groups of magnetic alloys one can distitinguish hard and soft magnetic materials (Hilzinger & Rodewald, 2013). At the moment a lot of new methods directed to creation of principally new methods of creation of new magnetic materials are being developed (Wang & Weng, 2016), for instance one can recall carbon (N. Li et al., 2018) and epoxy (Luo et al., 2018; Xiao et al., 2014) based composites, amorphous (Herzer, 2013; J. Zhang et al., 2012) materials, high enthropy magnetic alloys (P. Li et al., 2017) and heterostructures with enhanced magnetic properties (Jia et al., 2017). However the production of permanent magnets based on the metallic alloys is still relevant and has a high demand in production.

The Fe-Cr-Co ternary system is related to hard magnetic materials those magnetic properties are due to specific microstructure obtained during spinodal decomposition upon annealing. Normally this type of heat treatment is performed in the presence of the magnetic field allowing to obtain high values of remanence and coercivity however leading to the sharp drop of mechanical characteristics.

An important condition to achieve high physical, chemical, mechanical, technological and special properties is the optimization of the internal structure of the material. In particular, it is known that the magnetic and mechanical properties of materials are structurally sensitive and improving of required properties can be achieved by creating corresponding microstructure. The directed formation of structure of metals and alloys can be carried out using different methods of hot deformation, thus driving the processes of hardening and softening material in the process of dynamic recrystallization. Especially promising and important are the methods of severe plastic deformation, as in this case it is possible to obtain the structure of fine grained and in some cases even nanosized structure (Kawasaki et al., 2020; Zhilyaev & Langdon, 2008). Examples of technological schemes of realization of methods of severe plastic deformation (SPD) can be such processes of metals pressure treatment as longitudinal and transverse deformation, alternating bending, various pressing cross-helical rolling, and the combined circuit loading, including torsion, tension and upset. The latter allows (Akbar et al., 2014; Altafi et al., 2019; A. Korneva et al., 2019; Anna Korneva et al., 2019; Korznikov & Korznikova, 2017) to obtain the structure of gradient type with a nanostructured thin surface layer and a coarse-grained structure in the material volume. Knowledge of basic principles and temperature regimes of the dynamic recrystallisation normally taking place during the hot deformation process allows governing the microstructural parameters of the material in order to meet the technological requirements. However in this case one needs a high level of knowledge of the recrystallization dynamics aspects being pecular for every alloy in the ternary system.

Solving these problems requires construction of recrystallization diagrams, showing the dependence of the average grain size on the degree and temperature of the hot deformation at a given velocity. Recrystallization diagrams are the main reference information for specialists involved in design and development of new magnetic alloys with enhanced mechanical characteristics.

In the present work, diagrams of recrystallization for hard magnetic alloys Fe-25%Cr-15%Co and Fe-30%Cr-8%Co of the system Fe-Cr-Co are figured out basing on the results of experimental research of the microstructure of the material after the hot compression accompanied by torsion and revealing the dependencies of the microstructure on the deformation parameters.

2 .Materials and experimental details

Alloy billets of chemical composition Fe-25wt.%Cr-15wt.%Co and Fe-30wt.%Cr-8wt.%Co mentioned hereand after as 25X15K and 30X8K respectively were quenched from 1200°C in water in order to obtain the α-solid solution state. Cylindrical samples with a 12 mm diameter and 10 mm height were cut from the quenched billet. Samples were deformed at temperatures of 700, 750, 800 and 850°C, which corresponds to the single-phase α region in 30X8K alloy and two-phase $\alpha+\gamma$ region in the alloy 25X15K (Garganeev et al., 2018; Korneva et al., 2019; Korznikov & Korznikova, 2017). Combined deformation by compression and torsion was carried out on installation described in work (G. F. Korznikova & Korznikov, 2009), in isothermal conditions in two stages: the first stage for compression, and the second for torsion. The strain rate was varied in the range of $0.1\div0.01$ s^{-1}. The total logarithmic degree of strain was estimated using the approach given in (A. Korneva et al., 2013).

The microstructure of samples was studied in the scanning electron microscope JSM 6400 at an accelerating voltage of 10 kV. Microhardness was determined on the device PMT - 3 at a load of 0.2 kg according to the results of at least 15 measurements. Error of measurement on different samples, analyzed with the same degree of deformation was characterized by fluctuations within 5% range.

3. Results and discussion

Temperature - rate deformation conditions were chosen considering the range of superplasticity of alloys of the system Fe-Cr-Co. Samples after the deformation had no cracks and the microstructure along the cross section had a gradient type character.

Figure 1 shows a panoramic image and a detailed snapshots of alloys microstructure after combined deformation at a 750°C. As can be seen in Fig. 1 structure of specimens of both alloys in the cross section is strongly inhomogeneous with a noticeable gradient of grain size increase when moving from lower anvil to the upper one.

The distribution of the transverse grain size of of α-phase upon the sample cross-section of both alloys is shown in Figure 2. Analysis of grain size evolution shows that the value of grain size decreases with increasing temperature deformation in both alloys, and in two-phase alloy 25X15K one can distinguish three zones. The first zone corresponds to the region of intense deformation with the lowest grain size, the second zone is intermediate one and the third zone corresponds to the maximum grain size close to the pristine value. Thickness of the submicrocrystalline layer near the contact with the lower rotating anvil is 0,5 mm and weakly depends on the deformation temperature. In single-phase alloy 30X8K at temperatures of 700 and 750°C the grain size distribution regions are similar to that of the double phase one. Ultrafine-grained layer with grain size about 5 µm thickness is detected in the vicinity of the moving angle with the total thickness sbout 0.2 mm. In case of deformation temperatures of 800°C and 850°C grain remain coarse upon the entire cross section due to intensification of the dynamical recrystallization with temperature.

Measurements of microhardness in the cross section showed that the refinement of microstructure is accompanied by an increase of the microhardness. The distribution of microhardness for alloys 25X15K and 30X8K after strain loading by means of compression combined with torsion in isothermal conditions at 750 ºC along the sample cross section is presented in Figure 3. It is seen that deformation leads to an increase in Hv, however, the distribution of microhardness over the cross section of the samples is non homogeneous, and confirms the data of microstructural analysis results. Ultrafine grained regions in the active zone of deformation correspond to the highest values of microhardness for both alloys, with the total increase of values by 20% compared to the initial state.

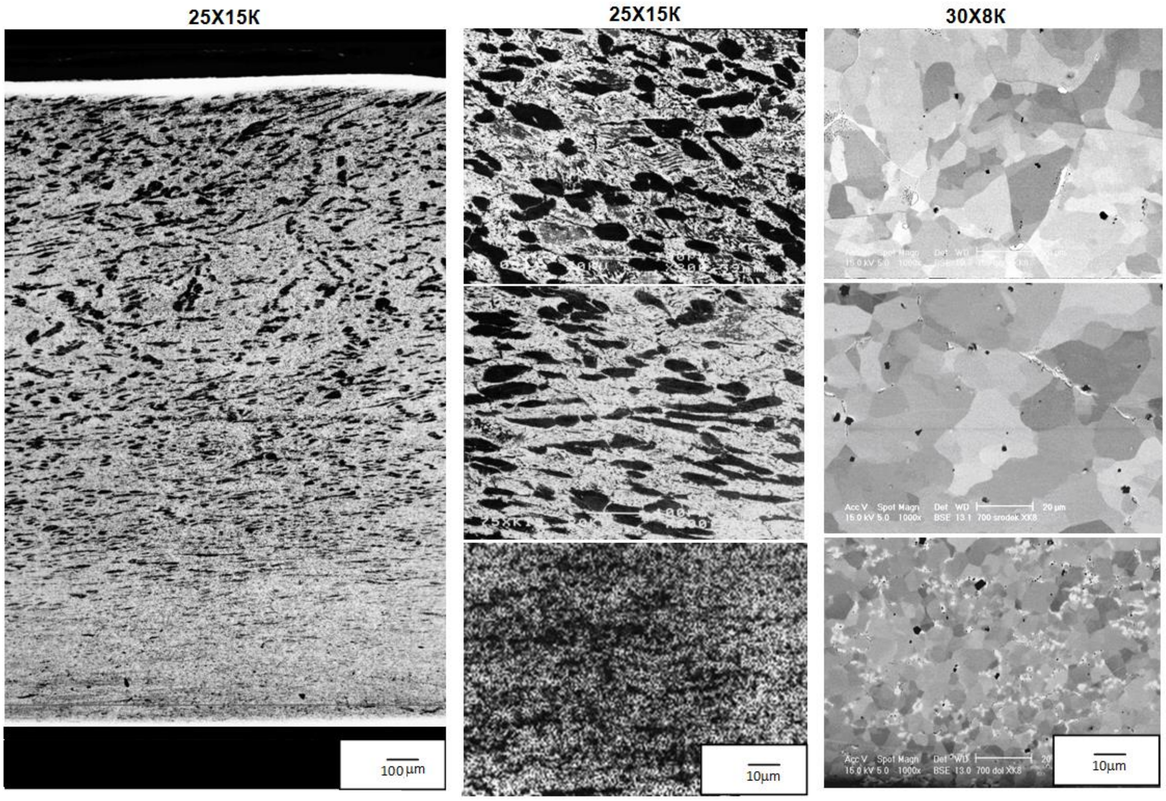


Figure 1. Panoramic image of the cross section of the 25X15K sample after complex loading after deformation at 750°C (left panel).Typical microstructure of the upper, middle, and lower parts of the samples of 25X15K and 30X8K alloys after deformation at 750°C presented on middle and right panels from top to bottom respectively.

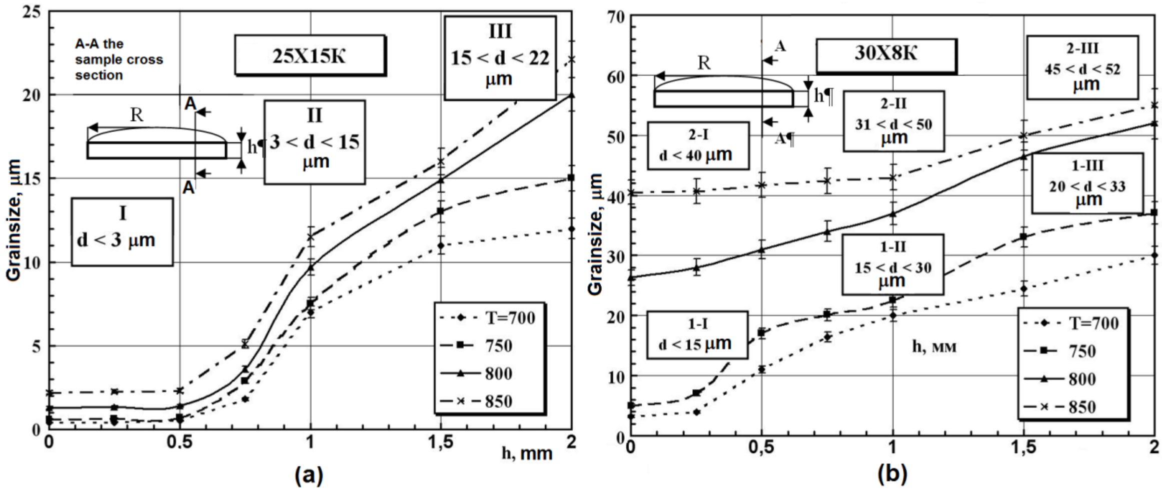
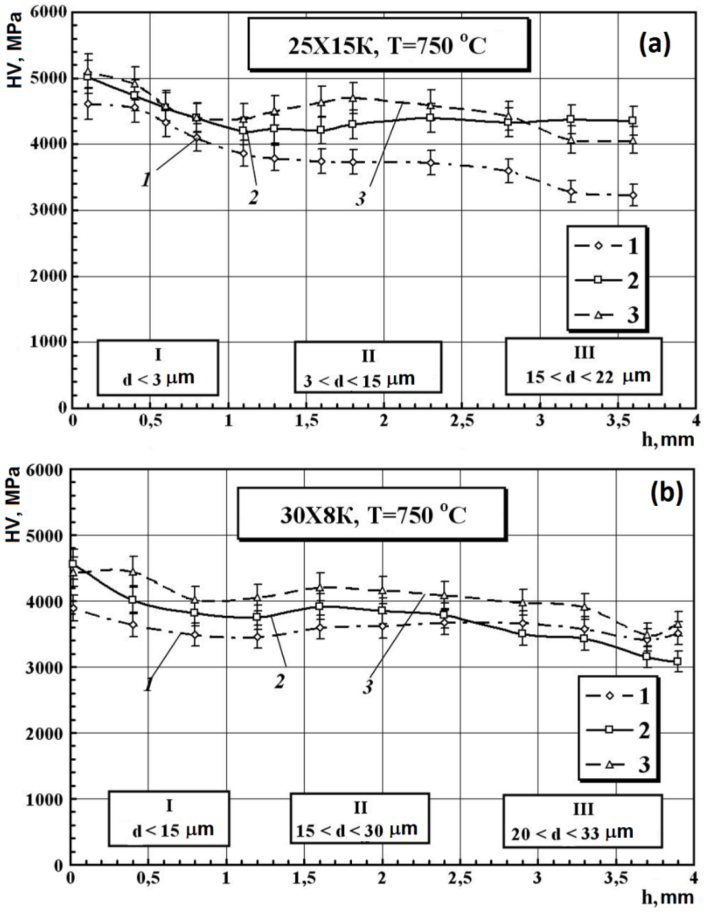


Figure 2. Grain size distribution over the sample cross section after complex loading at temperatures of 700°C, 750°C, 800°C, 850°C for 25X15K(a) and 30X8K (b) alloys.

One can observe the decrease of the microhardness value upon the increase of the distance from the active deformation zone in the vicinity of the lower mobile anvil. From Figure 3 it is clearly seen that the HV value measured along one layer on the distance R=0, R/2 and R from the rotation axis are about to be the same. One can also mention that the increase of the deformaiton temperature from 700 to 850 ºC leads to the reduction of HV in the zone of active deformation from 5400 to 4100 MPa and 4600 to 3200 MPa in the 25X15K alloy and in the 30X8K alloy respectively.



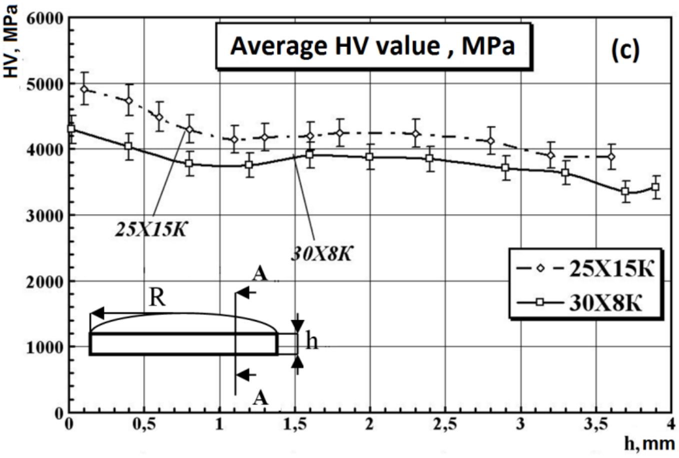
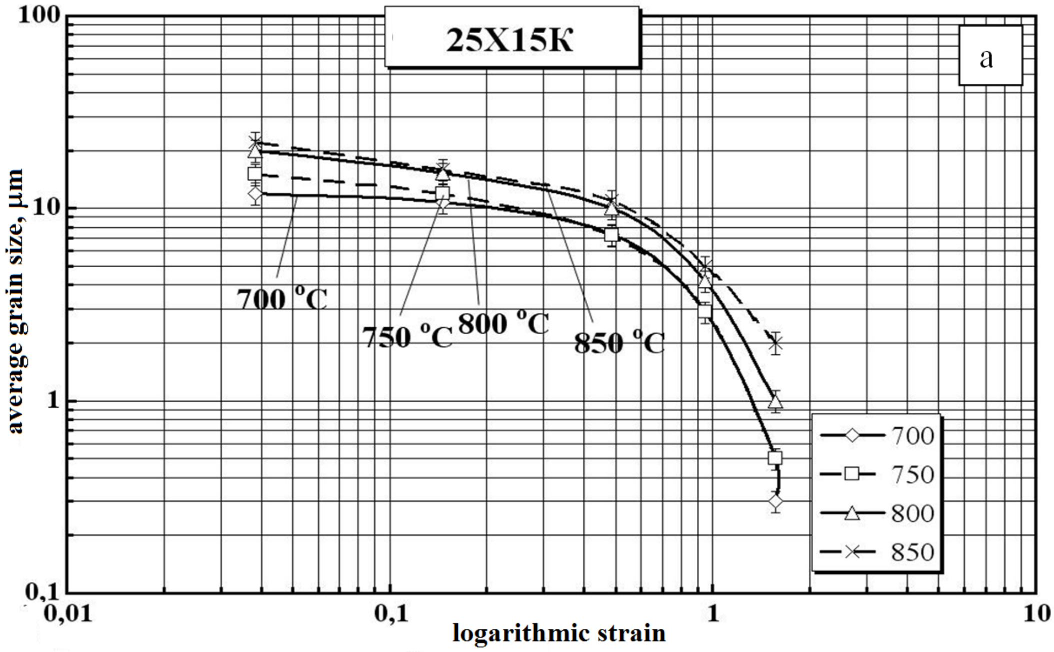


Figure 3. Change in microhardness along the height of the vertical section h of 25X15K (a) and 30X8K(b) alloy samples: 1 - in the center of the sample (R=0), 2 – at a distance R/2 from the center, 3 – along the side surface (R); (c) – average values of microhardness for both alloys.

The increase of microhardness after deformation in the temperature-rate conditions close to the regime of superplasticity, obviously, is primarily connected with the decrease of the grain size. In recent years, in physical mesomechanics has developed the approach based on accounting for the curvature of the crystal lattice (Panin et al., 2018). According to this concept of planar flows of structural transformations at the nanoscale level to form the curvature of the crystalline structure, which, in turn, leads to the development of plastic distortion of atoms from lattice sites to interstices of the areas of its curvature and the emergence of a large number of nonequilibrium vacancies. In the work (E. Korznikova et al., 2006) it is shown that the concentration of point defects (vacancies) in deformed materials is many orders greater than the equilibrium concentration. At elevated temperatures deformation can be expected growth of the contribution of point defects in the processes of evolution of structure, including their participation in appearance and motion of dislocation including both slide and climb options, and consequently, the strain localization associated with the development of the mechanisms of fracture of solids. In our case - in the conditions of low velocities and high temperatures of the deformation process the strain induced defects are emerged ontributing to relaxation of internal stresses associated with the lattice curvature. Therefore, the gradual formation of ultrafine-grained structure is observed instead of the sample fracture, and decrease temperature, similarly to the increase the deformation velocity, leads to the predominance of the accumulation of defects over their relaxation and, hence, a reduction in grain size and increase of microhardness.



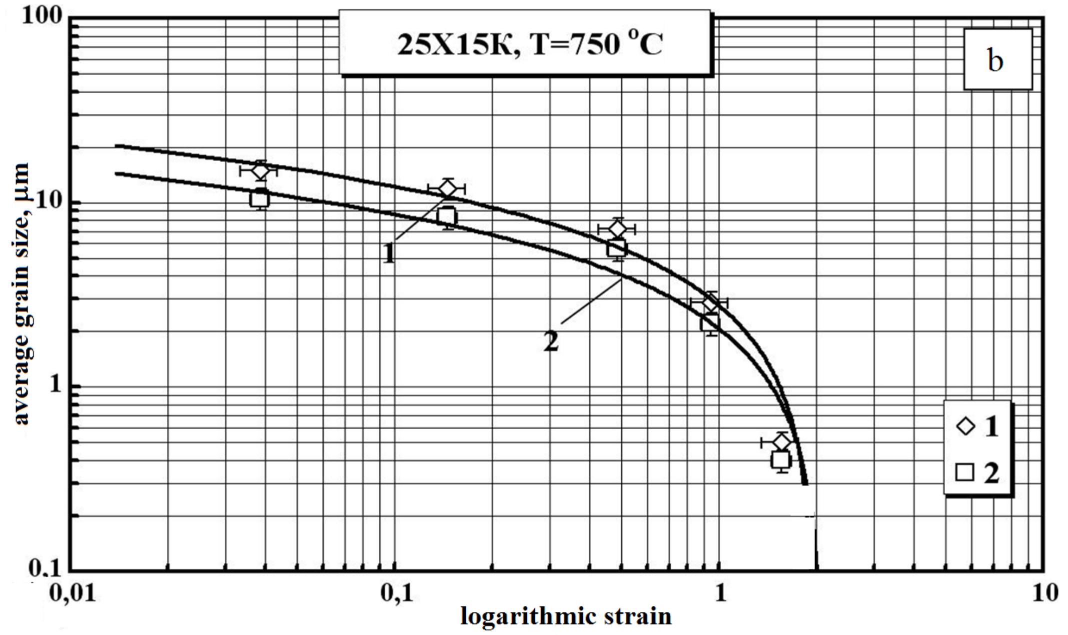
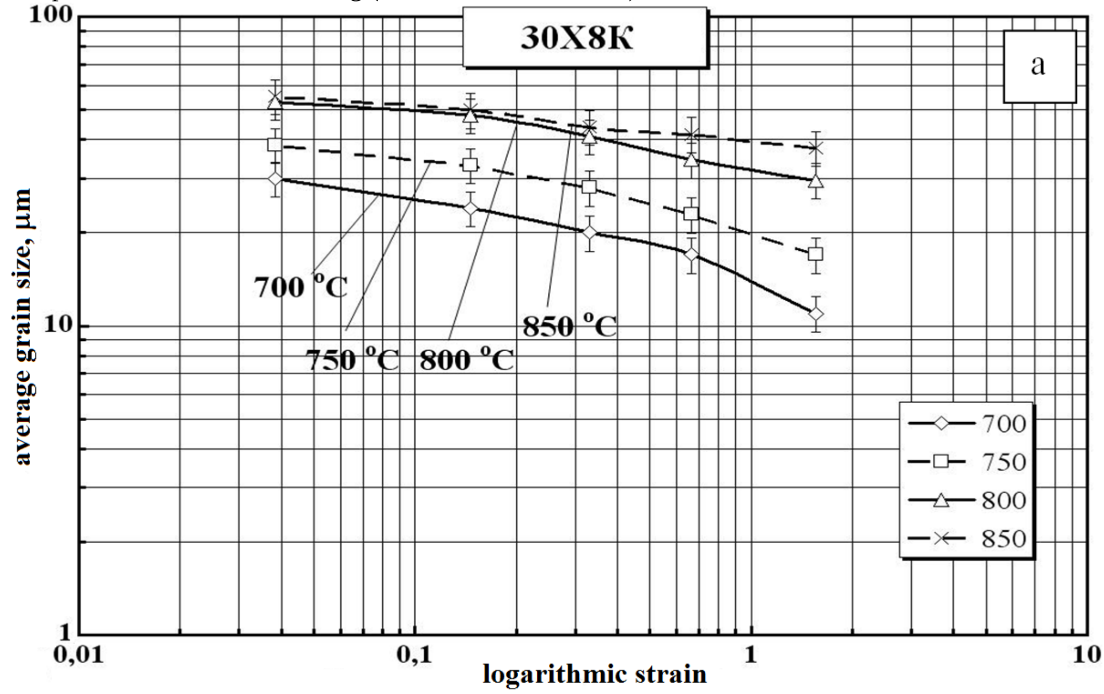


Figure 4. The dependence of the average grain size on the degree of deformation of the 25X15K alloy for different temperatures (a) and torsion rates (b) 1 -0,004 s^{-1}; 2 -0,017 s^{-1}.

The results of electron-microscopic studies of the samples processed–at different temperatures and deformation rates, allowed to elaborate recrystallization diagrams for the studied alloys 25X15K and 30X8K**.** Proper organization of technological processes of the production manufacture by different metal forming techniques is currently impossible without approved data on grain growth during processing. Grain growth is a major factor in various operations associated with hot processing of metals by pressure, inclu ding considered here schemes of severe plastic deformation. Recrystallization diagrams, which graphically depicts the relationship between grain size, degree of deformation, temperature and strain rate, are normally costructed for hot deformation cases (A. Korneva et al., 2019; Korznikov & Korznikova, 2017). These diagrams serve as guidelines for engineers and researchers during the processing of metals by pressure, though in their conventional form they can provide rather poor information on the nature of the recrystallized structure, and do not reflect many significant characteristics, for example, grain size distribution. Diagrams show the dependence of average grain size on the degree and temperature of hot deformation at a definite strain rate without post deformational annealing (A. Korneva et al., 2013).



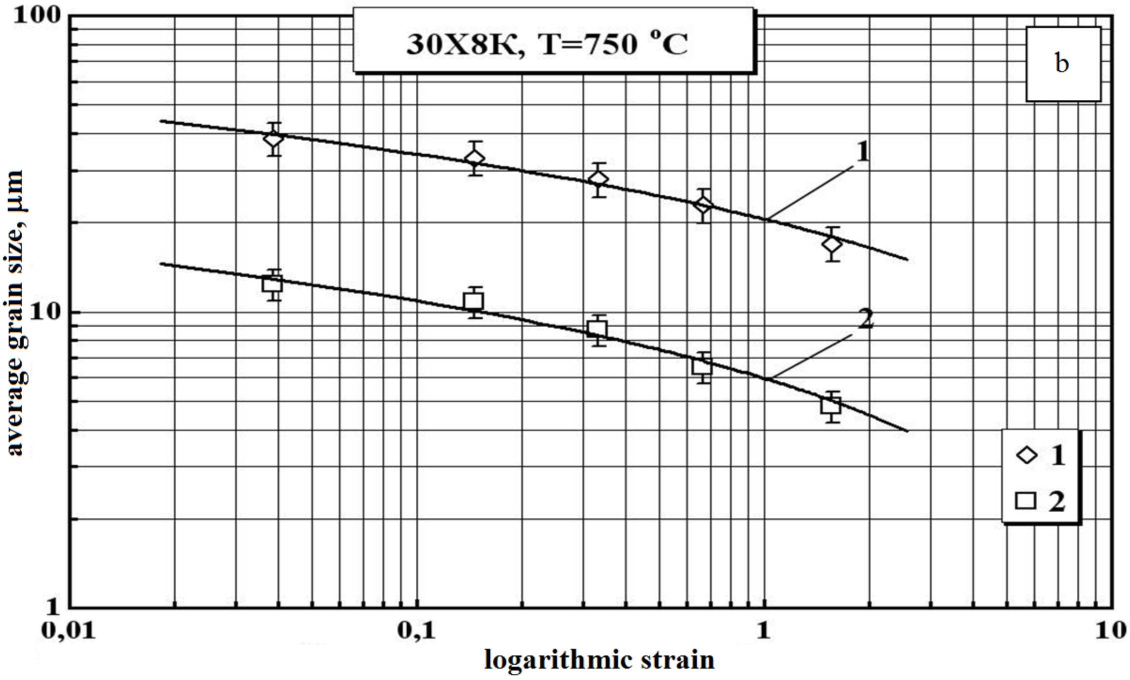


Figure 5. The dependence of the average grain size on the degree of deformation of the 30x8k alloy for different temperatures (a) and torsion rates (b). 1 - ω=0,25 rot/min, 2 - ω=1 rot/min.

The initial experimental data for elaborating recrystallization diagrams of the second order were the average grain size, strain temperature and strain rate obtained in the "torsion sediment" experiment, the calculated data was the degree of deformation (see (A. Korneva et al., 2019)). The calculation results are presented for 25X15K and 30X8K alloys in figures 4 and 5.

After processing the obtained graphic dependencies between the grain size, logarithmic degree of deformation, temperature and strain rate, it was found that for the 25X15K alloy, the grain refinement intensity increases by an order of magnitude when the logarithmic deformation reaches the total value more than 0.5. This is probably due to a change in the mechanism of dynamic recrystallization. The 30X8K alloy does not have such a change in the recrystallization mechanism, and the grain size reduction is not so significant.

In (G. F. Korznikova & Korznikov, 2009) the minimum grain sizes reached during hot Cr-Co alloys under complex loading conditions in two-phase 25X15K and single-phase 30X8K alloys were determined, and it was shown that the lowest level of structure refinement is limited by the spinodal decomposition phase field. Deformation at temperatures of spinodal decomposition leads to brittle fracture of the samples. In addition, it was found that with a decrease in the grain size of the α phase, the 30X8K alloy in the high-coercivity state exhibits higher mechanical properties. Therefore, the optimal grain size that provides the maximum level of mechanical properties properties is 0.5 microns for the 25X15K alloy and 5 microns for the 30X8K alloy. Based on the analysis, we can recommend using the following rational temperature and strain rate modes for processing by severe plastic deformation: for the alloy 25X15K 700÷900 °C и 0,1÷0,01 s^{-1}, and for 30X8K ~750±30 °C and 0,1÷0,01 s^{-1}

4. Conclusions

1. A comprehensive study of the processes of severe plastic deformation by means of compression accompained with torsion has shown that the chosen method of deformation can be effectively used for the structure refinement, which contributes to improving the strength characteristics of structural elements produced from 25X15K and 30X8K alloys.

2. Based on the analysis of research results, it is recommended to use the following rational temperature and strain rate modes for pressure treatment by severe plastic deformation of hard magnetic alloys: for 25X15K 700÷900°C and 0,1÷0,01 s^{-1}, and for 30X8K~750±30°C and 0,1÷0,01 s^{-1}.

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References

Akbar, S., Awan, M. S., Aleem, M. A., & Sarwar, M. N. (2014). Development of Mo Containing Fe-Cr-Co Permanent Magnets by Modified Single Step Thermomagnetic Treatment. *IEEE Transactions on Magnetics*, *50*(8). Scopus. https://doi.org/10.1109/TMAG.2014.2309433

Altafi, M., Ghasemi, A., & Sharifi, E. M. (2019). The influence of cold rolling and thermomagnetic treatment on the magnetic and mechanical properties of Fe-23Cr-9Co alloy. *Journal of Magnetism and Magnetic Materials*, *491*. Scopus. https://doi.org/10.1016/j.jmmm.2019.165537

Delette, G. (2015). Polycrystalline models of anisotropic sintered magnets: Influence of grain alignment on mechanical properties and residual stresses. *Journal of Magnetism and Magnetic Materials*, *389*, 10–20. https://doi.org/10.1016/j.jmmm.2015.04.016

Garganeev, A. G., Kyui, D. K., Kashin, E. I., & Sipaylova, N. Y. (2018). *Regulation Characteristics of Hysteresis Clutches Based on the Fe-Cr-Co Material*. 115–118. Scopus. https://doi.org/10.1109/APEIE.2018.8545340

Herzer, G. (2013). Modern soft magnets: Amorphous and nanocrystalline materials. *Acta Materialia*, *61*(3), 718–734. https://doi.org/10.1016/j.actamat.2012.10.040

Hilzinger, R., & Rodewald, W. (2013). *Magnetic materials: Fundamentals, products, properties, and applications*. Publicis ; VAC, VACUUMSCHMELZE.

Jia, X., Dai, R., Lian, D., Han, S., Wu, X., & Song, H. (2017). Facile synthesis and enhanced magnetic, photocatalytic properties of one-dimensional Ag@Fe3O4-TiO2. *Applied Surface Science*, *392*, 268–276. https://doi.org/10.1016/j.apsusc.2016.09.014

Kawasaki, M., Figueiredo, R. B., & Zhilyaev, A. P. (2020). Special Issue Celebrating Professor Terence G. Langdon's 80th Birthday. *Advanced Engineering Materials*, *22*(1), 1901386. https://doi.org/10.1002/adem.201901386

Korneva, A., Korznikova, G., Kashaev, R., & Straumal, B. (2019). Microstructure evolution and some properties of hard magnetic FeCr30Co8 alloy subjected to torsion combined with tension. *Materials*, *12*(18). Scopus. https://doi.org/10.3390/ma12183019

Korneva, A., Korznikova, G., Korznikov, A., & Sztwiertnia, K. (2013). Effect of deformation temperature on the microstructure of hard magnetic FeCr22Col5 alloy subjected to tension combined with torsion deformation modes. *Archives of Metallurgy and Materials*, *58*(2), 383–386. Scopus. https://doi.org/10.2478/amm-2013-0004

Korneva, Anna, Korznikova, G., Kashaev, R., & Straumal, B. (2019). Microstructure Evolution and Some Properties of Hard Magnetic FeCr30Co8 Alloy Subjected to Torsion Combined with Tension. *Materials*, *12*(18), 3019. https://doi.org/10.3390/ma12183019

Korznikov, A. V., & Korznikova, G. F. (2017). Influence of the structure on the deformation ability of the Fe-Cr-Co system alloys. *Materials Physics and Mechanics*, *33*(1), 104–112. Scopus.

Korznikova, E., Schafler, E., Steiner, G., & Zehetbauer, M. J. (2006). Measurements of vacancy type defects in SPD deformed Ni. *By YT Zhu, TG Langdon, Z. Horita, MJ Zehetbauer, SL Semiatin, TC Lowe.-Warrendate, PA: The Minerals, Metals & Materials Society (TMS)*, 97–102.

Korznikova, G. F., & Korznikov, A. V. (2009). Gradient submicrocrystalline structure in Fe–Cr–Co system hard magnetic alloys. *Materials Science and Engineering: A*, *503*(1–2), 99–102. https://doi.org/10.1016/j.msea.2007.12.059

Li, N., Jiang, H.-L., Wang, X., Wang, X., Xu, G., Zhang, B., Wang, L., Zhao, R.-S., & Lin, J.-M. (2018). Recent advances in graphene-based magnetic composites for magnetic solid-phase extraction. *TrAC Trends in Analytical Chemistry*, *102*, 60–74. https://doi.org/10.1016/j.trac.2018.01.009

Li, P., Wang, A., & Liu, C. T. (2017). A ductile high entropy alloy with attractive magnetic properties. *Journal of Alloys and Compounds*, *694*, 55–60. https://doi.org/10.1016/j.jallcom.2016.09.186

Luo, D., Wu, C., & Yan, M. (2018). Incorporation of the Fe3O4 and SiO2 nanoparticles in epoxy-modified silicone resin as the coating for soft magnetic composites with enhanced performance. *Journal of Magnetism and Magnetic Materials*, *452*, 5–9. https://doi.org/10.1016/j.jmmm.2017.12.005

Panin, V. E., Kuznetsov, P. V., & Rakhmatulina, T. V. (2018). Lattice Curvature and Mesoscopic Strain-Induced Defects as the Basis of Plastic Deformation in Ultrafine-Grained Metals. *Physical Mesomechanics*, *21*(5), 411–418. https://doi.org/10.1134/S1029959918050053

Wang, Y., & Weng, G. J. (2016). On Eshelby's S-tensor under various magneto-electro-elastic constitutive settings, and its application to multiferroic composites. *Journal of Micromechanics and Molecular Physics*, *01*(03n04), 1640002. https://doi.org/10.1142/S2424913016400026

Xiao, L., Sun, Y., Ding, C., Yang, L., & Yu, L. (2014). Annealing effects on magnetic properties and strength of organic-silicon epoxy resin-coated soft magnetic composites. *Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science*, *228*(12), 2049–2058. https://doi.org/10.1177/0954406213515112

Zhang, J., Chang, C., Wang, A., & Shen, B. (2012). Development of quaternary Fe-based bulk metallic glasses with high saturation magnetization above 1.6T. *Journal of Non-Crystalline Solids*, *358*(12–13), 1443–1446. https://doi.org/10.1016/j.jnoncrysol.2012.03.023

Zhang, Y., Zuo, T. T., Tang, Z., Gao, M. C., Dahmen, K. A., Liaw, P. K., & Lu, Z. P. (2014). Microstructures and properties of high-entropy alloys. *Progress in Materials Science*, *61*, 1–93. https://doi.org/10.1016/j.pmatsci.2013.10.001

Zhilyaev, A., & Langdon, T. (2008). Using high-pressure torsion for metal processing: Fundamentals and applications. *Progress in Materials Science*, *53*(6), 893–979. https://doi.org/10.1016/j.pmatsci.2008.03.002.